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Recent Advances in Adhesive Joining of Sustainable Materials

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Abstract

Nowadays, in order to increase the transport efficiency and reduce fuel consumption and emissions of pollutants, a reduction in weight associated with improved safety and performance of the materials in use must be achieved. On the other hand, environmental concern has generated interest in research about new materials aligned with the principles of sustainability. Therefore, this need for better performing and more ecological structures has resulted in the development of a new variety of materials. Among these materials, currently, are the composite materials produced from renewable sources. Natural fiber composites have recently attracted a great deal of attention by the industry due to their many attractive features (e.g., high strength-to-weight ratio, sustainable characteristics, and low cost). Welding is highly impractical to use in these situations (i.e., thermoset polymer composites) and rivets or screws lead to stress concentrations and offer a low fatigue resistance. Adhesive bonding is usually the preferred method to join these materials, since it allows for greater flexibility in design and is more efficient in mechanical and energy aspects. In this chapter, recent advances in adhesive joining of new sustainable materials, critical challenges, and future perspectives are presented. The application of sustainable composites may further increase in many structural and non-structural applications if their joining behavior is thoroughly understood and reliably proven.

***Keywords*:** Adhesive joints, composite materials, natural fibers, sustainable materials, mechanical characterization

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1.1 Introduction

Global initiatives and agreements, such as the United Nations Sustainable Development Goals (SDGs) and the Paris Agreement, have highlighted the urgency of adopting sustainable practices in all aspects of human activity. Industries are increasingly recognizing that sustainable materials not only align with ethical and environmental objectives but also offer economic advantages, including cost savings, enhanced brand reputation, and access to emerging markets with environmentally-conscious consumers. As a result, researchers, scientists, and engineers are actively engaged in developing innovative sustainable materials that exhibit improved performance characteristics while minimizing their environmental footprint.

Sustainable materials such as natural fiber reinforced composites (NFRCs) can be found in applications in many industrial sectors, such as: automotive, construction, marine, and sports, among others [1]. Moreover, the most promising applications of NFRCs are in the automotive industry. In general, they are used in non-structural car body parts, such as: door panels, package trays, hat racks, instrument panels, internal engine covers, sun visors, boot liners, etc. However, they are evolving to be used in more structurally demanding parts such as seat backs, exterior underfloor paneling as well as anti-roll bars [2–4]. They can also be used in construction, and building applications like bricks, door panels, roofing sheets, furniture panels, etc. The demand for natural fibers in polymer composites is expected to grow by 15–20% in the automotive sector and by 50% or more in certain building applications [5].

The main natural fibers studied and used in the industry (e.g., jute, sisal, kenaf, and flax) are well established on the global market with a well-defined production line. However, new promising types of natural fibers are being discovered and studied [1, 6]. Both thermoplastic and thermoset polymers are used as matrices in natural fiber reinforced polymer composites [7]. There is an increased interest in the scientific community in using bio-based polymers in composites as combining these matrices with natural fibers produces "green composites" or "bio-composites". An emerging trend is also found in the use of thermoplastic matrices of renewable sources, such as poly(lactic acid) (PLA), as a matrix material for NFRCs [8, 9]. This is crucial due to the fact that the classical thermoset matrix materials such as epoxy are dependent on fossil fuels.

Advanced bio-based materials that feature essential recyclability property, providing a sustainable alternative to conventional composites, are intensively investigated nowadays (see Figure 1.1). New bio-resins, new

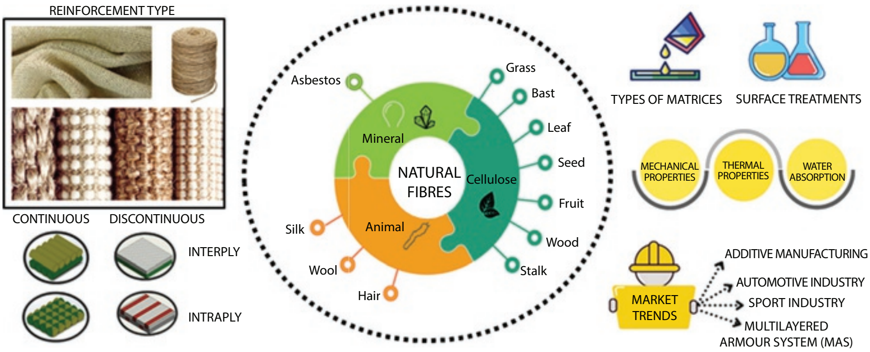

Figure 1.1 Schematic of development and characterization of new sustainable composites [1].

fibers, surface treatments of the fibers, the effect of water on the thermal and mechanical properties of new sustainable composites, different strategies to improve their properties are some of the topics that will be briefly covered in Section 1.2.

As the global emphasis on sustainability grows, the demand for more sustainable (eco-friendly) materials with enhanced properties has intensified. However, these sustainable composites must be joined in order to be used in industrial applications. There are many joining methods for connecting composites or composites to other materials (e.g. mechanical joining such as bolting, clamping or riveting). Moreover, it has been clearly demonstrated in the literature that adhesive bonding is the most favorable joining method for composites [10]. This chapter presents an overview of cutting-edge research and innovations in the development of more sustainable materials. It explores the challenges faced in adhesive bonding of these materials, as well as the innovative solutions and breakthroughs that have been achieved in recent years.

1.2 Development of New Sustainable Materials and the Main Strategies for Improving the Performance of These Materials

Sustainable materials are made from renewable, recycled, or waste resources, and can be recycled, biodegraded, or composted at the end of their life. These materials are designed to minimize environmental impact throughout their entire life cycle. This section will cover natural fiber reinforced composites with a focus on their mechanical properties and methods to enhance their performance (relevant for further using them in bonded structures). Extensive research is currently underway to enhance the mechanical properties and durability of these composites, aiming to make them competitive with traditional synthetic composites.

1.2.1 Natural Fiber-Reinforced Composites (NFRCs)

Recently, there has been an increasing growth in research and innovation in NFRC materials. NFRCs exhibit a wide range of mechanical properties. These properties, including tensile strength, flexural strength, and impact resistance, depend on various factors such as the type of fiber and matrix, the interfacial bonding between them, fiber dispersion, orientation, and processing methods. There is a common perception in academia and industry that bio-derived materials typically have relatively low performance and are suitable only for niche applications. Consequently, research in this field focuses on enhancing their mechanical capabilities, as by improving their performance, the potential applications of natural fiber-reinforced composites can be significantly expanded [11].

In order to explore the huge potential of replacing partially the synthetic fibers by natural fibers, many tests are required to explore their mechanical behavior. The most common mechanical tests used to characterize sustainable composites are the tensile, flexural and impact tests. Generally, the tensile strength of composites increases with fiber content up to a maximum value, then it tends to decrease. This is due to the higher mechanical properties of the fibers when compared to the matrices [2, 12]. Furthermore, it is well known that natural fibers contain moisture and when introduced into a hydrophobic matrix, the higher water absorption tendency of the natural fibers will lead to lower interfacial fiber/matrix adhesion and consequently lower mechanical properties of the NFRCs. One possibility to tackle this issue is the use of surface treatments [13]. The flexural properties of NFRCs are sensitive to a number of parameters, such as layup, ply angles, material type and stacking sequence [14]. This is due to the fact that the stresses increase from the neutral line and reach their maximum compressive and tensile stresses on the top and bottom of the test specimen, respectively. Impact resistance is a measure used to characterize the efficiency of the material during a rapid load event and is intrinsically linked to its toughness. The energy absorption may be derived *via* a penetration impact, residual properties after impact, and analysis of the damaged area after a non-penetration impact. Furthermore, the stacking sequence of the composite is a significant parameter when it comes to absorbed energy as it was observed that higher energy absorbing fiber layers on the outside of the composite enhance the overall impact resistance. On the other hand, the interfacial quality of the fiber/matrix is highly significant as a low-quality interface will poorly transfer the loads and decrease absorbed energy.

The main methods explored in the literature to increase the performance of natural fiber-reinforced composites are: fiber treatment and modification [15], the use of filler materials (either on the fiber surface or into the matrix as the second reinforcing phase) [16–18] and fiber hybridization [1].

Surface treatment of natural fibers is usually performed to enhance their mechanical and thermal properties, before using them to manufacture a composite material. The fiber modification techniques provide improved fiber-matrix interfacial adhesion, improved fiber roughness and wettability and depends on the particular fiber/matrix used and the composite application [15]. It is fundamental to understand the interfacial properties and bonding mechanisms of fiber-matrix and this requires significant research efforts in order to maximize natural fiber composites applications [13, 15, 19]. Another method explored in the literature to improve the properties of natural fiber-reinforced composites is the use of filler materials (either on the fiber surface or into the matrix as the second reinforcing phase) [16–18]. Due to their high specific surface area, good chemical interaction with the hydrophobic matrix and crack arresting capability, a synergistic relationship is observed with NFRCs [20]. However, a careful determination of the fabrication and matrix/filler homogenization technique must be done in order to avoid filler agglomeration, which may lead to detrimental effects. The most commonly used synthetic nanofillers are: Multi-walled carbon nanotubes (MWCNTs) and metal oxides.

One of the most popular methods to increase the mechanical performance of NFRCs to extend their applications is the fiber hybridization [1, 16, 21, 22]. Generally, natural fiber reinforced hybrid composites are produced by hybridizing natural fibers with another either natural or synthetic fiber with superior properties (e.g., high mechanical strength, chemical stability, nontoxicity, resistance to high temperatures, and thermal or acoustic insulation). The fiber hybridization techniques can be divided in: use of intralaminar hybridization, use of interlaminar hybridization, use of 3D fiber-reinforcement architecture (see Figure 1.2) and use of multi-scale hybridization.

For example, for the case of intralaminar hybridization, Pereira *et al.* [23] used ramie, sisal and curauá fibers as natural fiber reinforcements for the hybrid composites. They concluded that the mechanical properties are improved by the hybridization of sisal-based composites. On the

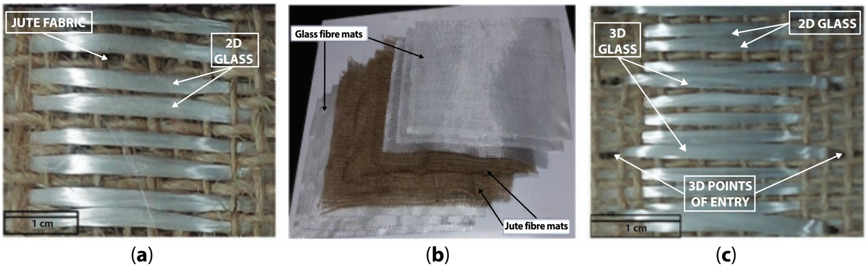

Figure 1.2 Types of architectural reinforcements: (a) intralaminar, (b) interlaminar and (c) 3D [11].

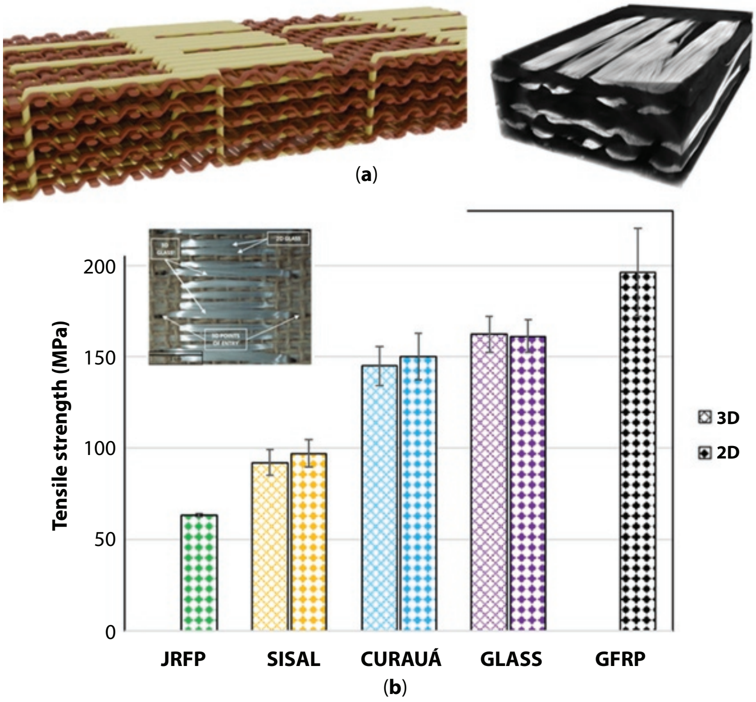

Figure 1.3 Example of use of 3D fibre-reinforcement architecture: idealized architecture and micro CTscan image (a) and effect of 3D reinforcement of hybrid natural composites (i.e., z-binder fibers stitched in the transverse direction through intralaminar 2D fabrics) vs. 2D reinforcement on the tensile stremgth (b). Where: GFRP—Glass fibre reinforced polymer composite; JFRP—Jute fibre reinforced polymer composite; SISAL 2D-Jute fabric reinforced with woven sisal UD; SISAL 3D-SISAL 2D reinforced with transverse orthogonal sisal fibres; CURAUÁ 2D-Jute fabric reinforced with woven curauá UD; CURAUÁ 3D-CURAUÁ 2D reinforced with transverse orthogonal curauá fibres; GLASS 2D-Jute fabric reinforced with woven glass UD; GLASS 3D-2D reinforced with transverse orthogonal glass fibres).

other hand, they state that the hybridization did not significantly affect the thermal stability of the composites studied. Interlaminarly hybridized composites of natural/synthetic fibers have been shown in the literature to present a non-linear enhancement of the material properties as the weight fraction of synthetic fibers increases.

de Queiroz *et al.* [24], demonstrated that for hybrid composites reinforced with a symmetric synthetic glass envelope around a jute fabric core, the in-plane properties increased significantly as the number of glass layers increased from 2 to 3 (i.e., an approx. 35% enhancement in tensile strength). This is due to the much higher density of synthetic fibers present in a woven fabric compared to the in-plane intralaminar hybridization. On the other hand, de Queiroz *et al.* [25] recently demonstrated that the effect of 3D reinforcement of hybrid natural composites (i.e., z-binder fibers stitched in the transverse direction through intralaminar 2D fabrics) is not significant in the in-plane properties (see Figure 1.3). The highest variation observed in the composite tensile strength (when the 2D case is compared to the 3D) was approx. 15%, while for most of the studied cases it was below 10%. Therefore, differently from the interlaminar reinforcement, significant variations would not be observed in the in-plane composite properties but rather in the transverse properties (strength and toughness).

1.2.2 Recent Advances in Development of NFRCs-3D Printing with Natural Fibers

One of the latest trends is the application of natural fibers as reinforcements in composites produced by additive manufacturing (AM or 3D printing). This technology allows for the fabrication of complex geometries without the need for expensive tooling and molds. It also allows for precise control over material distribution, opening up new possibilities for complex geometries and customizable structures [26]. Fused Deposition Modeling (FDM) commonly uses materials like acrylonitrile butadiene styrene (ABS) and poly(lactic acid) (PLA), but their inherent mechanical properties are limited. Incorporating nanoparticles and fibers into these materials can enhance their performance, making them suitable for high-performance applications [27]. These strategies aim to enhance the mechanical characteristics, thereby elevating the suitability of FDM-printed composites for applications demanding higher performance levels.

Recent research in this area focuses on different key aspects: studying the effects of filament type and printing parameters on the mechanical properties of AM parts [28], the mechanical performance of AM parts reinforced with natural (jute and curauá) and synthetic (glass) fibers [29], developing natural fiber/filler-reinforced PLA and ABS composites through fused deposition modelling [30]. For example, the use of natural fibers as filament reinforcement was investigated by Cavalcanti *et al.* [30] by using short curauá fibers with different lengths (3, 6, and 8 mm), and concentrations in terms of weight percentage (2, 3.5, and 5 wt. %) to fabricate poly(lactic acid) (PLA) filaments which were subsequently used to fabricate composites *via* fused deposition modelling (see Figure 1.4). They concluded that the curauá fiber-reinforced PLA composites may be a promising innovation to improve the performance of these materials, which might enable them to be used for new applications. However, some challenges remain to be solved in the fabrication of the reinforced filaments, such as difficult quality control due to voids and porosities, among others.

In another study [29], the mechanical properties of composite materials made by 3D printed (PLA and ABS) parts reinforced with different types of fibers were studied. Natural (jute and curauá) and synthetic (glass) fiber fabrics were laminated on the outer sides of the AM parts using an epoxy resin *via* compression moulding. The reinforcement was applied to the AM parts as 1 and 2-layers for each fiber type. The mechanical characterization

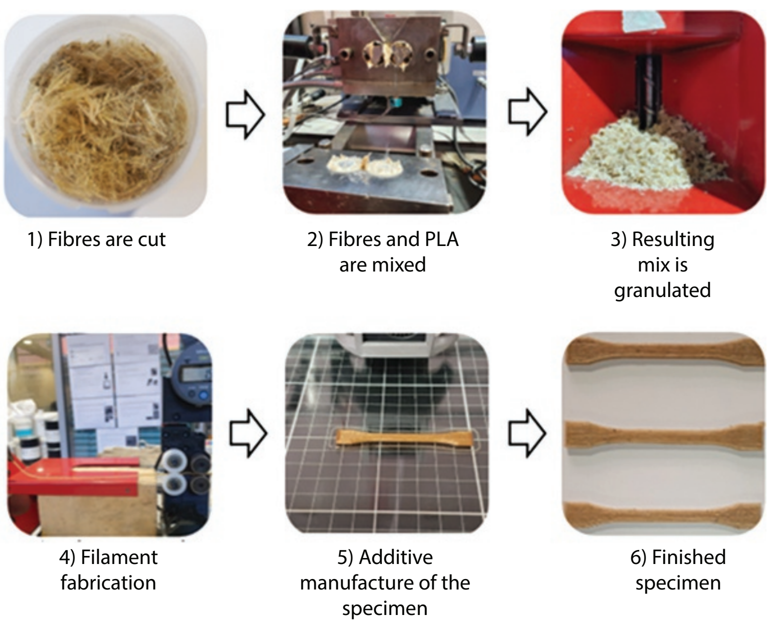

Figure 1.4 Schematic of the fabrication of the curauá fiber-reinforced PLA filaments [30].

of the resulting composites was performed through tensile and flexural tests in accordance with their respective ASTM standards. This work provides an alternative to the more common reinforcement methods, by using a hybrid manufacturing method: 3D printing+ compression moulding (see Figure 1.5). The application of novel manufacturing methodologies associated with renewable biodegradable fibers can provide an interesting path forward for fabricating high-performance 3D printed thermoplastic composites.

Enhancing the mechanical properties of additively manufactured parts through various types of fiber reinforcements is a promising innovation, as these improvements could enable these materials to be used in high load-bearing applications. However, due to the limitations of 3D printers in producing large parts, components often need to be printed separately and then assembled to create the final part. Additionally, 3D printing can be used to produce only the most complex parts, which can then be combined with simpler, non-printed components made from other materials to form the final product. One effective method for joining these additively manufactured parts is adhesive bonding. The behavior of adhesively bonded joints with 3D printed adherends was investigated by several researchers and was recently reviewed by Cavalcanti *et al.* [31].

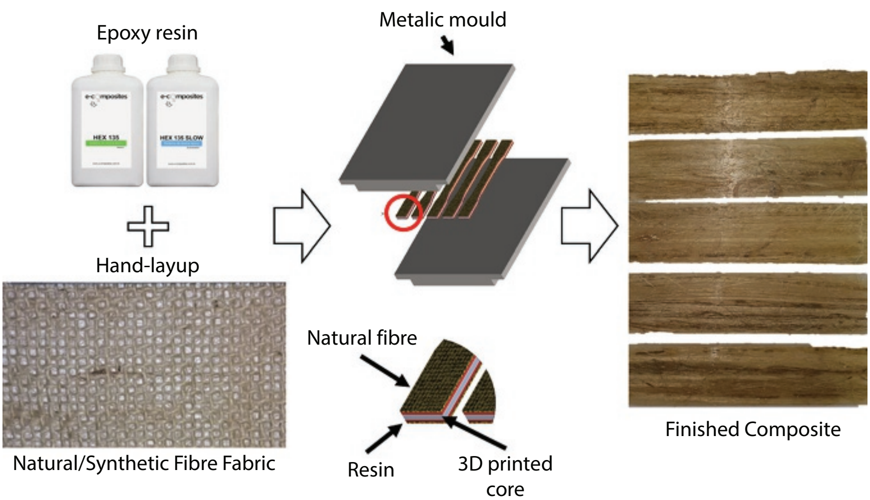

Figure 1.5 Schematic of the hybrid manufacturing method: 3D printing+ compression molding [11].

1.3 Adhesive Joining of New Sustainable Materials

The joining of new sustainable materials is of crucial importance in order to expand their applications [32, 33]. However, this field is still in its infancy and only little research has been done. One of the main issues facing the joining of sustainable materials is, for example, that natural fiber reinforced composites present significantly lower mechanical properties when compared to their synthetic counterparts. This is a significant drawback, and it hinders the bonded joint performance of these materials. Therefore, it is crucial to enhance the mechanical properties of sustainable materials prior to bonding them. The reason the material properties of composite are vital in a bonded joint is that due to the nature of the global geometry of the commonly used single lap joint (SLJ), an out-of-plane stress develops due to the eccentricity of the load path between the adherends (see Figure 1.6a). This peel stress is responsible for the crack initiation at the overlap edges where the stresses (both peel and shear) are highest. However, the propagation and path of the crack is controlled by other parameters, such as composite transverse strength and toughness. Furthermore, adherend stiffness will significantly impact its rotation tendency during the loading of the SLJ, either increasing or decreasing the peel stresses. This leads to the failure mode of delamination which is common for composite bonded joints and is characterized by the removal, partial or complete, of one or more layers of the composite (see Figure 1.6b).

Various techniques for reducing peel stresses within laminated adherends were examined in the literature [34]. However, there are many factors that affect the performance of the composite bonded joints [35]. The key parameters that influences the strength, durability, and overall effectiveness of the bonded joint are briefly summarized in Table 1.1. Among these, global design parameters—such as overlap length, bondline thickness, and fillet design—have the most significant impact on lap shear strength [36].

A number of researchers investigated the behaviour of adhesively bonded joints with sustainable materials and the main results are summarized in Table 1.2 [37–55]. Most of the works concentred on the performance of joints made of natural fibres-reinforced composites and methods to improve their performance mainly by tailoring the composite adherend properties. Adhesive joints of composites made from waste and of additive manufactured parts (mainly methods of enhancement of joint performance by optimizing the joint design and the surface properties of the adherends) were also studied.

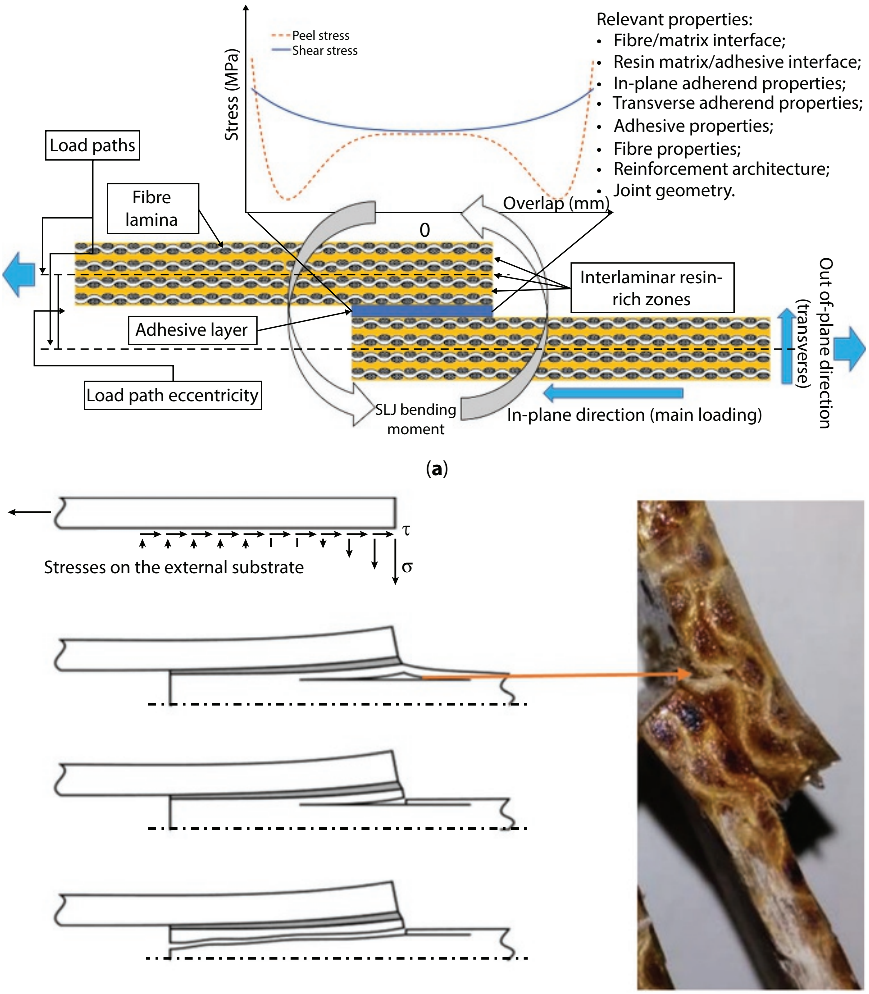

Figure 1.6 Schematic of composite bonded joint stress state and relevant parameters (a); examples of failure modes of NFRC bonded joint (b).

Several techniques exist to enhance the properties of adherends and adhesives, as well as to optimize local and global stress distributions. The primary goal of these methods is to eliminate delamination failures and maximize efficiency through effective joint design. Among these, the hybridization of NFRCs adherends has been the most studied and is found to be a successful technique. The hybridization technique has been shown to be the most effective in enhancing the material properties

Table 1.1 Key parameters that influence the performance of a bonded joint.

Parameters	Influence on joint performance
Adhesive properties	The adhesive material's mechanical properties, such as strength, stiffness, toughness, and durability, are critical factors that can affect the joint's overall performance. The compatibility of the adhesive with the composite material, including its resin matrix and fiber type, can significantly affect the bonding strength and durability of the joint.
Joint configuration	The choice of joint geometry depends on a variety of factors, such as the materials being joined, the loading conditions, and the desired performance characteristics. The joint configuration that produces local stress concentrations, high peel stresses and interfacial stresses should be avoided because it leads to premature failure of joints.
Overlap length	Increasing the overlap length generally enhances the load-carrying capacity of the joint by distributing the stress over a larger area. However, after a certain point, the benefits diminish, and additional length may lead to stress concentrations or increase the likelihood of adhesion failure.
Bondline thickness	An optimal bondline thickness is essential for effective stress distribution. A too thin bondline can lead to insufficient adhesive coverage and higher stress concentrations, while a too thick bondline can result in reduced joint strength and increased chances of adhesion failure due to poor load transfer.
Adherend Thickness	The thickness of the adherends affects the joint's stiffness and its ability to distribute stress. Thicker adherends typically improve joint strength by reducing peel and shear stresses, but they can also lead to a more brittle failure mode. In contrast, thinner adherends may allow for more flexibility but might suffer from increased deformation and reduced load-bearing capacity.

(*Continued*)

Table 1.1 Key parameters that influence the performance of a bonded joint. (*Continued*)

Parameters	Influence on joint performance
Adhesive Fillet Geometry and Taper Angle (Adherend End Geometry)	The shape and size of the adhesive fillet at the edges of the joint influence stress distribution and the prevention of peel stresses. A well-formed adhesive fillet can reduce stress concentrations at the joint edges and improve the joint's resistance to peeling and shear forces, thereby enhancing joint strength and durability. The geometry of the adherend ends impacts the stress distribution near the edges. A well-designed taper reduces stress concentrations, at the ends of the overlap, and enhances the joint's overall fatigue life and load-bearing capacity. Sharp edges or abrupt changes in geometry can significantly increase stress concentrations and reduce joint durability.
Adherend Surface Preparation and Roughness	The surface roughness and treatment of the adherend influence the adhesive's ability to bond effectively with the adherend. Proper surface roughness increases the effective bonding area, leading to better load transfer and joint performance.
Manufacturing method	The application of pressure during bonding, curing conditions, and surface preparation all play crucial roles. Properly controlled manufacturing methods ensure strong, uniform bonds with minimal defects, leading to better load transfer, reduced stress concentrations, and enhanced resistance to fatigue and environmental degradation. Conversely, poorly executed bonding can result in weak joints, increased susceptibility to failure, and reduced overall structural integrity.

of these more environmentally-friendly materials, usually by employing a small weight fraction of synthetic fibers. This hybridization is done in the form of distinct architectural techniques, such as: intralaminar, interlaminar, 3D architectures and multi-scale reinforcements (see Figure 1.7). The intralaminar reinforcement architecture is defined by the application of two or more reinforcement fiber phases within the same fabric layer (see Figure 1.2a). This architecture is more complex and labor intensive,

Table 1.2 Research efforts in adhesively bonded joints with sustainable materials.

Adhesive	Adherend	Main findings	Refs.
Epoxy, polyurethane, and PLA (parent polymer of composites)	bamboo/ PLA green composite	The width of the specimen and type of adhesive were the most significant parameters affecting both tensile and compressive failure load. The bonded joints failed due to adhesion failure, cohesive failure and fiber-tear failure.	[50]
Bostik 7452 (Rubber and Plastics Grade) ethyl cyanoacrylate	hemp fibre/ polypropylene composite	Polypropylene/hemp natural fibre layer adjacent to the bonded joints were compared to single fibre composites. The strength of joint was higher in case of single fibre composite. The fatigue strength of hybrid stacked joints was lower than the single fibre composite joints due to lower adhesion between hemp and adhesive layer.	[37]
Epoxy	henequen and sisal fibre/epoxy composite	Three different geometry types: single lap shear joints (SLJs), co-cured joints termed intermingled fibre joins (IFJs) and laminated fibre joints (LFJs) were studied. The IFJs and LFJs presented higher lap shear strengths than the SLJs.	[38]
Epoxy	jute fibre/epoxy composite	The overlap lengths and curing temperatures in both single and double lap joints were varied. The delamination is the dominant type of failure in both single lap and double lap joints.	[39]
Epoxy	kenaf fibre/epoxy composite	Single lap hybrid joints tests and numerical modelling. Good agreement between experimental and numerical results.	[40]

(*Continued*)

Table 1.2 Research efforts in adhesively bonded joints with sustainable materials. (*Continued*)

Adhesive	Adherend	Main findings	Refs.
Epoxy	bamboo natural fibre/polyester composite	The effect of position of bamboo natural fibre layer on the failure strength of the joint was studied. The position of the reinforcement had an effect on the stress concentration of the adhesive joint.	[51]
Epoxy	jute fibres/ interlaminar jute-glass fibre outer layers/epoxy composites	Significant joint failure load was observed as a function of hybridization. Optimum load was found for 2 glass layers (i.e., 100% failure load of the fully synthetic glass fiber-reinforced polymer composite (GFRP) joint.	[24]
Epoxy film EA9686 STRUCTIL	flax/epoxy composites	The impact of surface roughness, induced by sandblasting, in Mode-I. The Mode-I fracture energy decreases with the increasing of the roughness mean depth R.	[44]
Acrylic	flax/epoxy composite	The influence of surface treatments, like sandblasting, on the shear strength of bonded joints was studied. The rougher surfaces enhanced bond strength in Mode II.	[45]
Epoxy	jute and sisal fibre intralaminar hybridization with sisal, curaua and glass/ epoxy resin composites	Significant joint failure load enhancement was found as a function of hybridization. The highest bonded joint performance improvement was found for the jute/glass intralaminar hybrid specimens (the hybrid joint reached approx. 77% of maximum joint load efficiency of the GFRP synthetic joint).	[4]

(*Continued*)

Table 1.2 Research efforts in adhesively bonded joints with sustainable materials. (*Continued*)

Adhesive	Adherend	Main findings	Refs.
Epoxy	short curauá fibres and glass fibre bidirectional fabrics/ epoxy resin composites	The failure mode of bonded joints is heavily impacted by the composition and arrangement of the composite layers in the adherends. The number of synthetic layers had a significant effect on the bonded joint's failure load, whereas the symmetry of the architecture showed no notable impact	[52]
Epoxy	jute and carbon fibres/ epoxy resin composites	Hybridization significantly enhances SLJ joint performance, achieving up to 97% of the strength of carbon fiber-reinforced composites (CFRPs) while also offering a more sustainable and cost-effective alternative to fully synthetic composites.	[53]
Epoxy	jute and interlaminar hybrid jute-glass fibres/ epoxy resin composites	Adhesive joints with hybrid composites demonstrate higher shear fracture energy (G_{IIC}) compared to joints made solely with synthetic fibres. This improvement in fracture toughness is due to the synergistic interaction between the natural and synthetic fibres.	[54]
Epoxy	jute and curauá/ PLA and ABS composites	The mechanical performance of adhesively bonded SLJs was significantly enhanced by incorporating various types of fiber reinforcements into the 3D-printed core adherends. The failure load is influenced by the type of fiber, the number of reinforcement layers, and the material used for the adherend cores.	[55]

(*Continued*)

Table 1.2 Research efforts in adhesively bonded joints with sustainable materials. (*Continued*)

Adhesive	Adherend	Main findings	Refs.
Epoxy	PLA composite	The effect of sinusoidal tailoring of the overlap coupled with different building directions (flat wise and edge wise). A good degree of compatibility between the PLA adherends and the epoxy adhesive used was found and all joints presented failure in the adherends.	[27]

but a highly tailored composite architecture can be achieved. On the other hand, the interlaminar reinforcement architecture is simpler, where each layer is comprised of a single fiber type, usually a fabric (see Figure 1.2b). This arrangement has the advantage of being able to leverage the synthetic fiber's high water barrier property to keep moisture from entering the composite. This is particularly relevant for natural fibers which have a high hydrophilic tendency. Another crucial material property of a composite to be bonded is the transverse strength and toughness. Due to the much lower transverse strength, the delamination failure is directly linked to the peel stresses exceeding the strength and a crack initiating and propagating in the interlaminar space. Therefore, the technique of 3D reinforcement comes into play to solve this issue by applying a z-binder fiber phase that binds the fabric layers in the transverse direction, significantly increasing the composite transverse strength and delamination toughness (see Figure 1.2c).

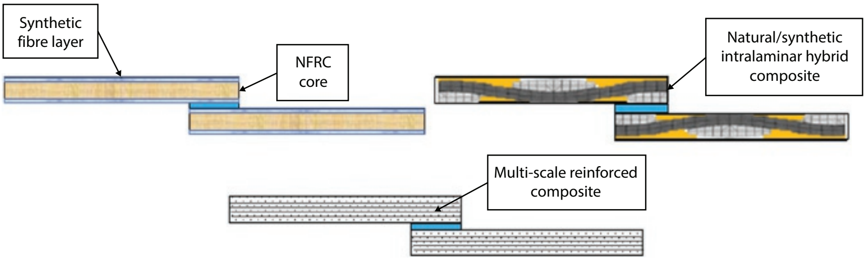

Figure 1.7 Schematics of the main methods used to increase the robustness of natural fibre-reinforced composite (NFRC) adhesive joints.

The effect of intralaminar reinforcement in the bonded joint performance of hybrid composites was analyzed by de Queiroz *et al.* [4]. Jute and sisal fabrics were used as base fiber phases for intralaminar reinforcement *via* weaving with glass fiber to fabricate hybrid composites. Neat jute, sisal and glass fiber reinforced composites were also fabricated. A two-component epoxy matrix was used to fabricate the composites using the compression molding. Single lap joints, both similar and dissimilar, were fabricated and tested in quasi-static tension. It was found that it is possible to reach up to approx. 77% of the bonded joint failure load of the neat GFRP *via* the glass fiber intralaminar hybridization of jute fabrics. In a follow-up study by de Queiroz *et al.* [24], the interlaminar reinforcement technique was employed to enhance the bonded joint performance of jute/glass fiber composites. Glass and jute fiber fabrics were used, with the glass layers positioned on the exterior of a 5-layer jute fiber core. The number of exterior glass fiber layers varied between 2 and 3, arranged symmetrically. An epoxy matrix, along with a structural automotive two-component epoxy adhesive, was used to fabricate the composites and single lap joints. It was found that a hybrid interlaminar glass/jute composite with 3 glass layers achieved approximately 100% of the bonded joint failure load of a fully synthetic glass fiber reinforced polymer composite (GFRP), as can be seen in Figure 1.8. This performance was attributed to the optimal balance of improved adherend material properties. Finally, in a recent research

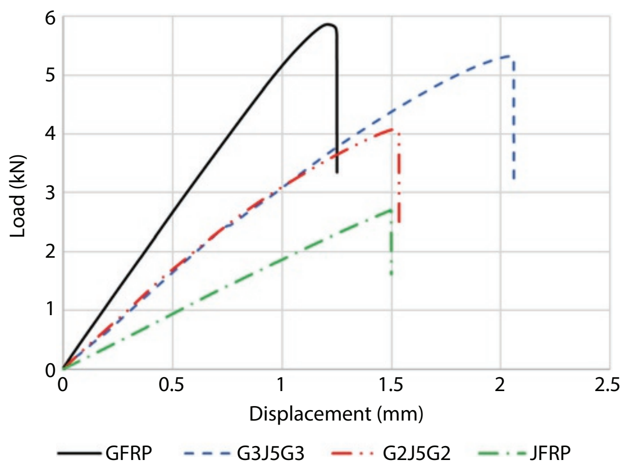

Figure 1.8 Representative load-displacement curves of SLJs as a function of adherend material (where: GFRP—Glass fibre reinforced polymer composite; G2J5G2—2-layer glass fibre hybrid composite; G3J5G3—3-layer glass fibre hybrid composite; JFRP—Jute fibre reinforced polymer composite) [24].

effort, de Queiroz *et al.* [25], investigated the effect of a novel 3D reinforcement architecture in the bonded joint performance of hybrid composites. Bidirectional jute fabrics were used as well as sisal, curauá, hemp and glass fibers in order to fabricate intralaminarly reinforced 2D fabrics. These fabrics were used to fabricate 3D fiber-reinforced preforms by weaving a

Table 1.3 Summary of data from recent research efforts in adhesively bonded joints of sustainable materials [4, 24, 25, 52, 53].

Adherend material	Failure load (kN)	Displacement (mm)
NEAT CURAUÁ (short fibre curauá reinforced composite)	2.56±0.20	1.5±0.37
JFRP (neat jute fibre reinforced polymer composite)	2.89±0.19	1.88±0.34
*GLASS A2 (short curauá fibre core, 2 glass bidirectional layers one side)	3.46±0.29	1.5±0.2
*GFRJP (intralaminar glass + jute bidirectional)	4.24±0.22	1.51±0.15
*G3J5G3 (interlaminar glass (3 layers)+5 layers of jute reinforcement)	5.53±0.18	2.74±0.83
*AC2 (asymmetric carbon bidirectional fabric reinforcement-2 layers)	5.64±0.22	2.04±0.29
SISAL 3D (Intralaminar sisal reinforcement of jute fabric + 3D orthogonal sisal reinforcement)	5.30±0.15	2.22±0.11
CURAUÁ 2D (intralaminar curauá reinforcement of jute fabric)	5.47±0.21	1.95±0.5
CURAUÁ 3D (CURAUÁ 2D reinforced with transverse orthogonal curauá fibres)	5.51±0.26	2.18±0.11
GFRP (neat glass fibre reinforced polymer composite)	5.51±0.27	1.53±0.29
CFRP (neat carbon fibre reinforced polymer composite)	5.61±0.35	1.25±0.30

**natural/synthetic hybrid composites*

z-binder fiber through 2D fabrics. The composites were fabricated *via* the compression molding technique. Single lap joints were fabricated with a modern automotive grade epoxy adhesive. It was found that the 3D reinforcement successfully enhanced the bonded joint performance of the novel sustainable composite materials for both the sisal and curauá specimens. This also marks the first time in the literature where a fully natural fiber reinforced composite joint matched approx. 100% the bonded joint performance of a fully synthetic joint (i.e., GFRP and CFRP).

For comparison, typical values of failure load and displacement for SLJs with sustainable adherends found in literature are summarized in Table 1.3.

1.4 Summary and Future Perspectives

The increasing importance of sustainable materials is a response to the pressing need to address global environmental challenges. This background sets the stage for a deeper exploration of recent advances in the development, characterization, and joining of new sustainable materials, as industries seek transformative solutions to create a more sustainable and resilient future. This chapter outlines recent advances in developing and joining sustainable materials, with the main focus on natural fiber composites. Several approaches exist to improve joint performance, including geometrical optimizations, adherend modifications, and adhesive modifications. Among these, hybridization of NFRC adherends has emerged as the most successful technique, demonstrating significant improvements in bonded joint efficiency while maintaining relatively low fabrication complexity. However, while hybridization shows promise, other methods—such as wavy overlap geometries, fillets/tapers, and 3D reinforcements—also offer potential but require further research and validation.

Several promising research directions and emerging trends could further advance the field of adhesive joining of sustainable composite materials:

- Fully sustainable structures: The development of fully biodegradable composite systems, where both the fibers and the matrix—including the adhesive—are biodegradable, is a critical area for future research. Developing new adhesive formulations to further improve the performance and sustainability of new materials and innovations in adhesive formulations that facilitate easier disassembly and recycling of components at the end of their lifecycle will be essential for promoting sustainability.

- Optimized joint designs and fabrication: Future research should explore functionally graded joints with varying adhesive/adherend properties to optimize stress distribution. Tailored joint architectures, such as scarf, stepped, or hybrid joints, can also be explored to maximize mechanical performance in sustainable materials. Additionally, there is significant potential for further enhancing the geometrical optimization of bonded joints. New optimization methods tailored for specific applications could lead to more efficient joint designs with improved performance. The integration of additive manufacturing techniques, such as 3D printing, in fabricating adhesive joints with complex geometries and precisely applied adhesives can allow for customized joint designs with enhanced strength and reduced material waste.
- Advanced computational models and standardization: The use of finite element analysis (FEA) and other advanced computational tools will play a vital role in predicting the behavior of adhesive joints under various loading conditions. These models can help optimize design parameters, leading to better-performing joints. There is still a need for industry-wide standards to ensure consistent performance evaluation and reliability of adhesive joints involving new sustainable materials.

As the demand for sustainable and recyclable materials continuously grows, the application of NFRCs is expected to expand, particularly in the automotive, construction, and sports industries. Continued research into hybridization, fabrication methods and other advanced joining techniques will be crucial for meeting the increasing performance requirements of these applications.

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